



Dynamics of nitrogen, phosphorus, and organic pollutant losses from a small watershed in the drinking-water source protection area in Guiyang City of Southern China

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Abstract

Nutrients in runoff degrade water quality. The development of schemes to mitigate such degradation requires a characterization of the underlying dynamic processes of nutrient loss. The drinking-water source protection area in the Lake Hongfeng watershed of Guiyang City, the capital of Guizhou Province, China, has been delimited for effective conservation. However, no systematic observations have provided data on nutrient losses from these areas that could support optimal management. We selected one typical watershed in the area. Automatic gauges were installed to record the water levels and calculate runoff rates during 2010 and 2011. A total of 1523 runoff samples were collected at an interval of 3 h during a day; total nitrogen (TN), total phosphorus (TP), and chemical oxygen demand (COD) were tested. The results indicated that surface runoff rates were primarily less than 15 L/s but rapidly increased 1–30 times 15 L/s when it rained. TN, TP, and COD concentrations primarily fluctuated between 0.06 and 18.79 mg/L, between 0.01 and 1.57 mg/L, and between 0.01 and 160 mg/L, respectively. TN and COD concentrations in 98.98% and 52.04% of the runoff samples, respectively, exceeded the upper limit required by the Environmental Quality Standards for Surface Water (EQSSW) in China. Conversely, 94.29% of the runoff samples had lower concentrations than the upper limit of TP concentration. Surface runoff has been seriously polluted by nitrogen and organic pollutants. The occurrence frequency of different runoff rates and TP and COD concentrations showed different distributions, but TN concentrations had a normal distribution. There was a significant relationship between runoff rates and TP concentration and TN, TP, or COD loss. TN, TP, and COD loss primarily occurred on vegetable lands, rice fields, and residential sites. Effectively controlling nitrogen fertilizer that is applied on vegetable lands and paddy fields and managing wastewater and solid waste are urgent. The results reported here will also provide references for many other regions facing similar problems.

Keywords Nonpoint source pollution · Runoff · Nitrogen · Phosphorus · Water quality

Introduction

Environmentalists found that water quality in rivers and bays was not significantly improved after point source pollution

had been effectively controlled during the 1960s; the water quality of these rivers and bays even continued to deteriorate (Wang et al. 2011). This phenomenon represents a new type of diffusive pollution, i.e., nonpoint source pollution different

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from point source pollution. Therefore, the nonpoint source pollution became a large concern. Currently, N and P losses have significantly increased compared with those in prior decades (Holzmann et al. 2016). The continually increasing levels of N and P fertilization are unlikely to cease in developing countries, which will cause more serious nonpoint source pollution (Wesström et al. 2014). In China, the use of nitrogen and phosphorous fertilizers and pesticides has approached the limit. The superfluous use of nitrogen and phosphorous fertilizers has caused eutrophication in almost all fresh lakes and rivers and has impacted drinking water quality (Zhang et al. 2004).

To effectively manage a watershed to control nonpoint source pollution, many researchers have attempted to characterize temporal distributions, transport loads, and dominant pathways for N and P loss and to develop prediction models (Ferreira et al. 2018). For example, Alberts et al. (1978) determined the seasonal loss of N and P in surface runoff from three agricultural watersheds located in southwestern Iowa. A long-term study indicated that the flow-weighted nitrate nitrogen concentrations during an 11-year period averaged 13.4 mg/L and 12.0 mg/L on two pieces of land used to grow corn with conventional tillage and no tillage, respectively, but the mean annual subsurface drain flow was 35 mm higher for no tillage (315 mm) than conventional tillage (280 mm) (Randall and Iragavarapu 1995). Zhang et al. (2007) observed up to ten times greater N and P concentrations in runoff from three forested watersheds in central Japan than in base flow and reported that the majority of total nitrogen (TN) and total phosphorus (TP) loss in runoff was discharged from April to June. In a 50.1-ha watershed in which more than 80% of land was paddy fields, TN and TP loads ranged from 1.7 to 3.9 kg/ha and from 0.15 to 0.77 kg/ha, respectively, during growing seasons (Kim et al. 2006). For an assessment of nitrogen and phosphorus loss from croplands or watersheds, many prediction models, such as the storage treatment overflow runoff model, agricultural nonpoint pollution source, and soil and water assessment tool, have also been established (Young 1989; Neitsch et al. 2001). Compared with other parts of the world, in China, systematic studies of the quantification of the temporal distribution, transport loads, and dominant pathways for N and P loss and the development of prediction models for achieving sustainable watersheds and preserving the environment are relatively rare (Fu 2018).

Environmentalists are especially concerned with N, P, and chemical oxygen demand (COD) loss from the watersheds of Lake Tai, Lake Cao, Lake Dongting, Lake Dianchi, and Lake Boyang, i.e., five crucial lakes, in terms of determining underlying mechanisms and optimal management of nonpoint source pollution in China (Zhu et al. 2018). However, high-frequency observations have never been conducted to determine the export dynamics of nonpoint source pollutants in the crucial Lake Hongfeng watershed in a karstic region for the

purpose of effective management. Previously, some ecologists only estimated the loads of nonpoint source pollution from the whole watershed and the drinking-water source protection area of Lake Hongfeng (Yu et al. 2012; Geng et al. 2017). Lake Hongfeng is the largest freshwater lake in Guizhou Province, China, which provides 70% (4×10^5 m³/day) of the drinking water for Guiyang City, the capital of Guizhou Province. Guiyang City has more than four million people. Water pollution in Lake Hongfeng directly threatens drinking water safety in Guiyang City. There is substantial intensive agriculture with excessively applied fertilizers in the drinking-water source protection area of Lake Hongfeng (Yang et al. 2016). Model predictions showed that loads of nonpoint source pollution from the drinking-water source protection areas were high and caused serious water pollution and eutrophication of the lake (Geng et al. 2017). It is necessary to monitor N and P loss from the drinking-water source protection area of Lake Hongfeng to specifically identify the pollution risk to ensure the development of suitable conservation policy. The implementation of ecological engineering for controls of nonpoint source pollution also requires the support of actual observational data.

In the field, observers often monitor runoff rates and nutrient concentrations, and then, nutrient loads during a temporal interval such as a day, month, or year may be calculated by multiplying the runoff rates and corresponding nutrient concentrations. However, runoff rates and nutrient concentrations are not always sampled with the same frequency due to limitations of techniques. Thus, through simple regression approaches, relationships between nutrient loads and runoff rates have been developed to estimate transported nutrient loads (Alexander et al. 1996; Tiemeyer et al. 2006). For example, power functions are often used for the estimation of TN and TP loss with runoff (Kim et al. 2006; Fujii et al. 2006; Tomer et al. 2003). However, the occurrence frequency of different runoff rates and nutrient concentrations in runoff caused by random rainfall events has not been studied. Occurrence frequency can also be applied to determine the probability distribution of different runoff rates and nutrient concentrations with a goal of watershed management. Understanding the occurrence frequency can also improve representativeness in the sampling of different runoff rates and nutrient concentrations in the field. For example, if the occurrence frequency of a certain level of runoff rates or nutrient concentrations is relatively higher than that of another level, the level of runoff rates or nutrient concentrations may be considered to collect relatively more samples.

The Lake Hongfeng watershed includes many subwatersheds. We selected a representative small watershed, the small Dachong watershed, within the Lake Hongfeng watershed. In the Dachong watershed, there are different types of land uses, and the agricultural production level and population density are also representative. We monitored runoff rates and

N and P concentrations in surface runoff at the exit of the small Dachong watershed during 2010 and 2011 and quantified the temporal distribution and output loads of these indices. These results were also compared with those obtained from nonkarstic regions in Asia, Europe, and Africa. In these regions, crop types and the production levels of agriculture or rainfall are similar to those in the Dachong watershed. However, these regions are not part of drinking-water source protection areas. This comparison clarifies the levels of water pollution in the Dachong watershed at a global scale and even in the whole Lake Hongfeng watershed. We primarily answer the following questions: (1) What are the dynamics of runoff rates and N and P concentrations based on each observation and daily or monthly averages? (2) What types of probability distributions do the occurrences of different runoff rates and N and P concentrations in runoff belong to? (3) How much of the total loss of surface runoff, TN, TP, or COD has occurred from the small Dachong watershed comparatively with watersheds in nonkarstic regions and does the total loss (or loads) explain the roles of different types of land uses in the small Dachong watershed? (4) What relationships are there between runoff rates and N, P, and COD concentrations and loss with runoff?

Materials and methods

The small Dachong watershed

The small Dachong watershed (105° 55′–106° 30′ E, 26° 07′–26° 39′ N) is located approximately 45 km away from the center of Guiyang City, the capital of Guizhou Province, China. Lake Hongfeng (52.7 km² of water surface) is the primary source of drinking water for Guiyang City. A section with a 2 km width around the lake is delimited as the drinking-water source protection area in which farming is dominant. The small Dachong watershed is distributed within the protection area. Although there are many watersheds in the protection area, the upper section of these watersheds is usually distributed outside the protection area. The small Dachong watershed is also a representative of the Lake Hongfeng watershed on the Yungui Plateau, where karst is widely distributed. The area of the watershed is 1.239 km², and the altitude ranges from 1105 to 1250 m. The surface runoff from the watershed directly flows into Lake Hongfeng, one drinking water source for Guiyang City. The watershed is characterized by a subtropical climate. The annual mean temperature is 14 °C, and the annual mean rainfall is 1360 mm, 80% of which occurs between June and October. The types of landforms include hills, flat lands, and a broad valley. Hills are covered by a secondary evergreen broadleaf forest (SEBF), sparse young forest (SYF, this type includes orchards), and Chinese fir plantation (CFP) (Vogel et al. 2011). Slope cropland (SC) and terraced fields (TFs)

are distributed at the bottom of hills. Vegetable lands (VLs) and paddy fields (PFs) are distributed on flat land along the rivers (Fig. 1, Table 1). Primary soil types are yellow soil and paddy soil. There is no exposed limestone or sinkholes, but there is a permanent fountain. In the watershed, 620 people primarily live on croplands, and there are no factories. For a long time, large quantities of nitrogen fertilizers have been applied to croplands to maintain high crop yields, which have actually exceeded crop requirements (Xu et al. 2012). The TN contents in the soil of PFs, TFs, and VLs were higher than the values of the highest (or the most fertile) grade of agricultural soil defined by the *Classification Standard of Soil Fertility in China* (Table 2).

Observation of water level and calculation of runoff quantity

A rectangular thin-plate weir was installed in the main stream at the basin outlet (Fig. 2). The basin outlet is approximately 50 m away from Lake Hongfeng and 10 m higher than the water surface of the lake. In addition to the weir, we built a well; the water at the bottom of the weir was able to flow into the well through a culvert. A water level logger (Onset HOBO) was installed in the well with an overhanging line. The pressures (P , Pa) under water with changes in the water table were recorded by the logger at an interval of 10 min (Fig. 3). The water level (h , m) at the moment when the pressures under water were recorded was calculated using Eq. 1. The runoff rate (Q , m³/s) was given using Eq. 2 (Ministry of Urban and Rural Construction 1993). We also installed an automatic meteorological station. Rainfall, atmospheric pressure on the ground surface, wind speed, and air moisture were automatically observed.

$$P - P_0 = \rho gh \quad (1)$$

$$Q = 0.018 B h^{3/2} \quad (2)$$

where P_0 (Pa) is the atmospheric pressure on the surface of the ground at the basin outlet, which was determined based on observed values; ρ is the water density (10³ kg/m³); g is the gravitational acceleration (9.8 m/s²); and B is the width of the weir (m).

Rainfall in the middle section of the watershed was observed by a self-recording rain gauge (HOBO RG3) installed on the roof of a building (approximately 15 m in height). The observed results were checked using the data collected from the automatic weather station.

Collection and test of water samples

The collection of water samples was conducted at an interval of 3 h from 8:00 in the morning to 20:00 in

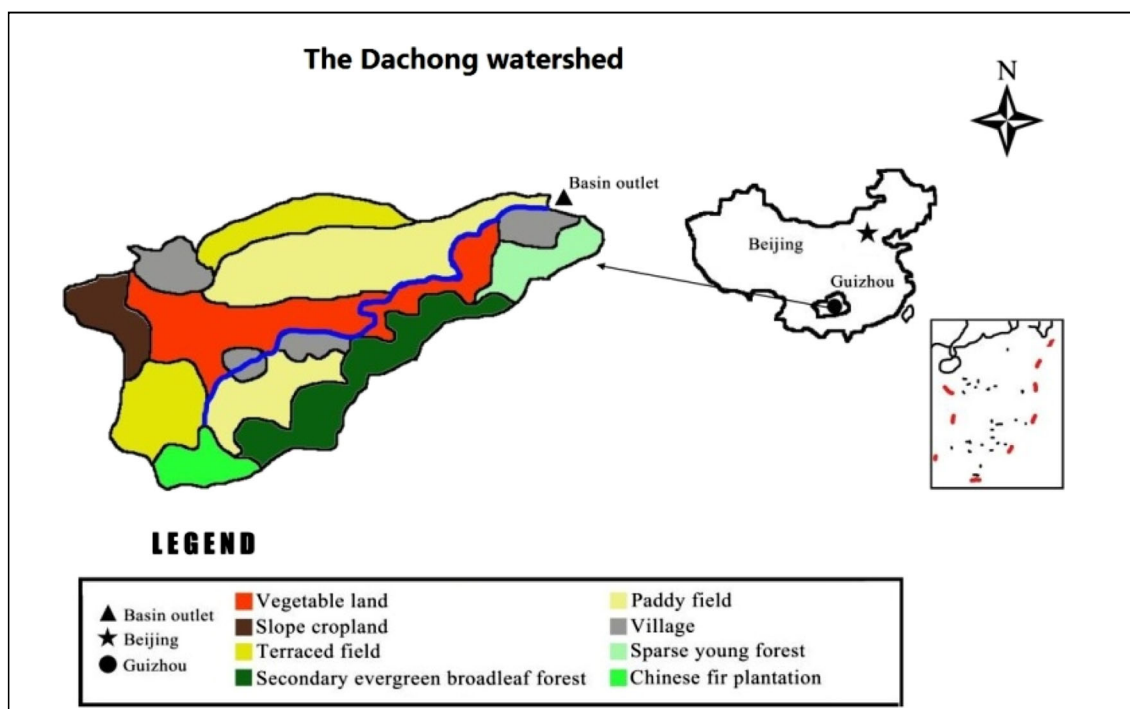


Fig. 1 Location of the small Dachong watershed

the evening during each day in 2010 and 2011. On the 15th day and 30th day of each month, water samples were also collected during the night from 20:00 in the evening to 8:00 in the morning of the second day. We assumed that the number of water samples collected and tested at all these moments over the 2 years would be able to represent the water quality of surface runoff from the small watershed. When it was time to collect water samples, approximately 500 mL of runoff was taken with a water sample collector from the channel of the rectangular weir. Then, the collected water was transferred to a 500-mL plastic bottle cleaned in advance and kept at 2 °C in the laboratory approximately 800 m away from the rectangular weir in the small

Dachong watershed. TN, TP, and COD in each bottled water sample without any mixing were measured with alkaline potassium persulfate digestion UV spectrophotometry, the ammonium persulfate digestion spectrophotometric ammonium molybdate method, and dichromate titration, respectively (Ministry of Environmental Protection of China 2002a).

Data analysis

The dynamics of runoff rates and TN, TP, or COD concentrations in runoff samples over the 2 years are shown in the figures. The different intervals between the maximum and minimum runoff rates and TN, TP,

Table 1 Land use in the small Dachong watershed

Land use types	Area (ha)	Percentage (%)	Mean slope (°)
Vegetable land	23.20	18.7	0
Slope cropland	6.30	5.1	14.03
Terraced field	12.50	10.1	0
Paddy field	25.10	20.3	0
Sparse young forest	10.10	8.2	15.36
Chinese fir plantation	3.80	3.1	14.03
Secondary evergreen broadleaf forest	28.00	22.5	16.73
Other lands*	14.90	12.0	N/A

N/A not applicable

*Other lands include residential areas, roads, and water body

Table 2 Soil nutrient status of different types of land use in the small Dachong watershed

Land use	Bulk density (g/cm ³)	Organic matter (g/kg)	Total nitrogen (g/kg)	Alkaline hydrolyzable nitrogen (mg/kg)	Nitrate nitrogen (mg/kg)	Total phosphorus (g/kg)	Available phosphorus (mg/kg)
Vegetable land	1.19 ± 0.01	43.21 ± 0.65	2.65 ± 0.02	295.75 ± 2.54	20.15 ± 1.22	0.67 ± 0.01	7.52 ± 0.31
Slope cropland	1.11 ± 0.03	29.32 ± 0.14	1.66 ± 0.03	92.34 ± 1.37	10.27 ± 0.36	0.60 ± 0.03	7.52 ± 0.18
Terraced field	1.23 ± 0.03	31.17 ± 0.23	2.15 ± 0.03	239.28 ± 1.29	18.07 ± 1.65	0.93 ± 0.01	9.02 ± 0.15
Paddy field	0.98 ± 0.01	58.74 ± 0.10	3.31 ± 0.04	506.55 ± 1.44	1.04 ± 0.12	0.73 ± 0.01	9.01 ± 0.12
Sparse young forest	1.57 ± 0.01	30.54 ± 0.13	1.56 ± 0.01	104.61 ± 1.07	1.12 ± 0.06	0.23 ± 0.02	6.65 ± 0.17
Chinese fir plantation	1.49 ± 0.01	77.65 ± 1.08	2.60 ± 0.05	167.37 ± 1.23	5.08 ± 1.23	0.49 ± 0.02	9.45 ± 0.13
Secondary evergreen broadleaf forest	1.16 ± 0.01	42.65 ± 0.19	1.88 ± 0.02	125.20 ± 2.56	3.07 ± 0.29	0.35 ± 0.05	3.32 ± 0.14

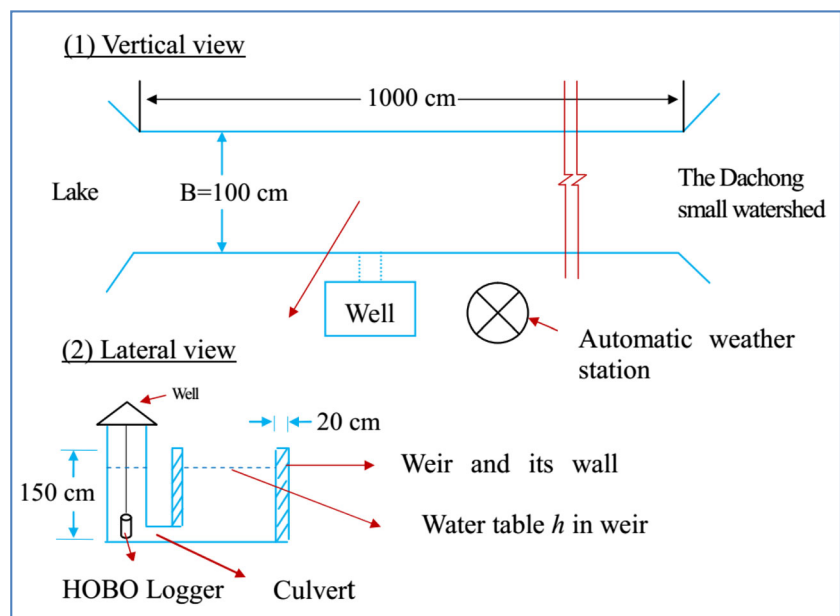
or COD concentrations were determined. The occurrence frequency of the runoff rates and TN, TP, or COD concentrations falling into each interval was counted. The probability distributions of different runoff rates and TN, TP, or COD concentrations in 2010 and 2011 are presented in histogram figures. The Kolmogorov-Smirnov method was further used with IBM SPSS 19 to test and identify whether their distributions belonged to normal, uniform, exponential, or Poisson distributions. TN, TP, or COD loss with runoff between two neighboring runoff samples collected from the weir was calculated by multiplying the average of TN, TP, or COD concentration in two neighboring runoff samples with corresponding runoff quantity (Zhang et al. 2005; Chun et al. 2010). Relationships between TN, TP, and COD concentrations or loss and runoff rates were quantified by regression models.

Results

Rainfall

The rainfall in 2010 was 1157.1 mm, and 84.66% of the rainfall occurred from April to September (Fig. 4a). There were 154 rainfall events recorded throughout the year, including 117 rainfall events < 10 mm, 22 rainfall events between 10 and 25 mm, 8 rainfall events between 25 and 50 mm, and 7 rainfall events between 50 and 100 mm. The rainfall in 2011 was only 688.6 mm, and the year 2011 was obviously an extreme drought year. Only 64.54% of the rainfall occurred from April to September compared with that in 2010 (Fig. 4b). There were 116 rainfall events recorded throughout the year, including 103 rainfall events < 10 mm, 8 rainfall events between 10 and 25 mm, 2 rainfall events between 25 and 50 mm, and 3 rainfall events between 50 and 100 mm (Supplemental

Fig. 2 Rectangular thin-plate weir



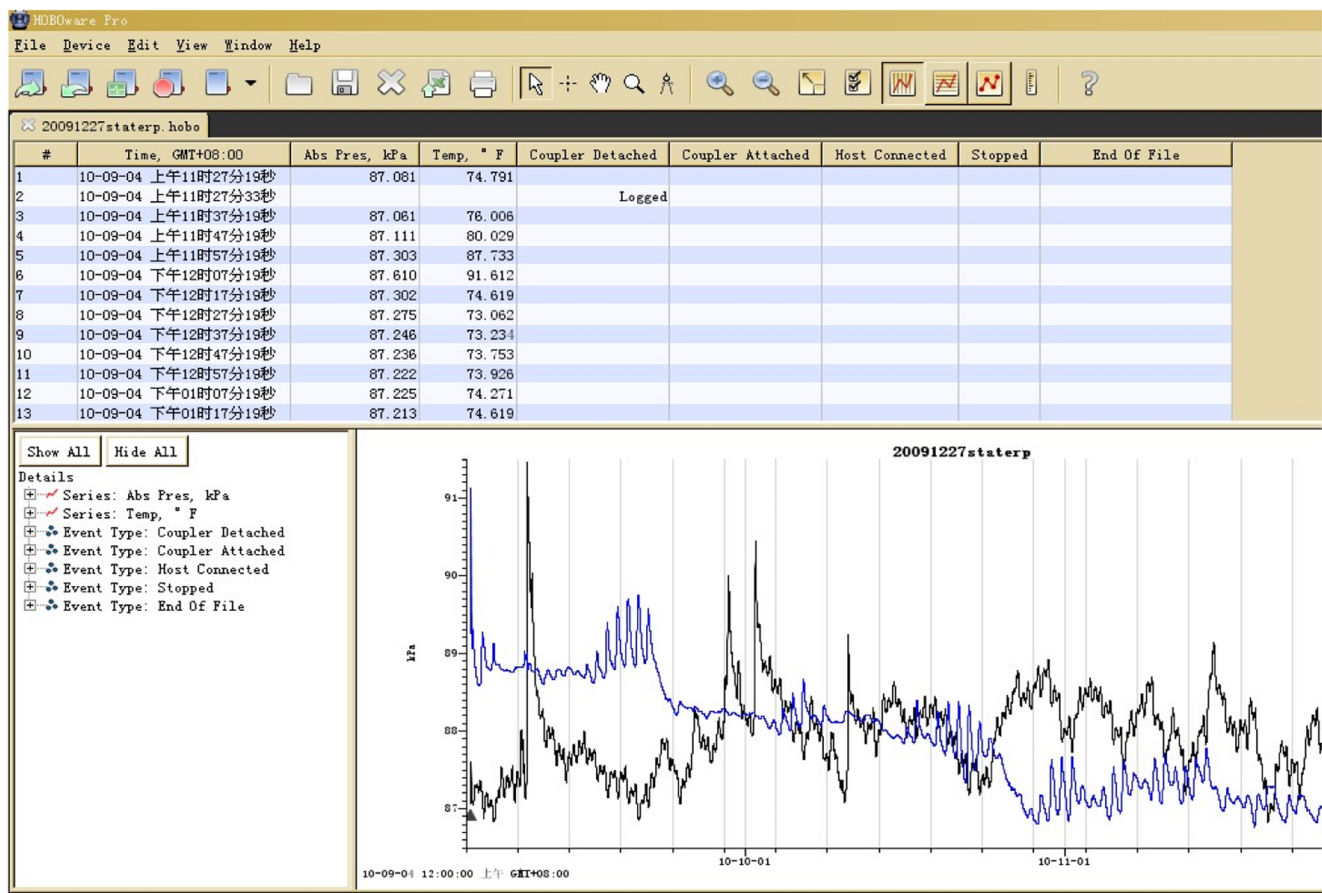


Fig. 3 Data recorded by the low-energy water level logger

material 1). The seasonal variation between the 2 years was very significant (F value = 3.22, p = 0.024, and n = 12, ANOVA).

Runoff rates

Runoff events occurred at the rectangular weir for a total of 211 days in 2010. These runoff events primarily occurred between April and September (Fig. 5a(1)). The maximum and minimum runoff rates were 705.88 L/s and 0.19 L/s,

respectively. However, runoff events only occurred at the rectangular weir for 84 days in 2011. The runoff events primarily occurred between May and July and during October (Fig. 5a(2)). The maximum and minimum runoff rates were 788.80 L/s and 0.18 L/s, respectively. The maximum runoff in the normal rainfall year 2010 was smaller than that in the drought year 2011. All observed runoff rates and the daily average runoff rates in both 2010 and 2011 fluctuated greatly, but the peaks and valleys of the runoff rates in 2010 were more variable than in 2011 (Fig. 5a(1), b(1), a(2), b(2)). The

Fig. 4 Monthly rainfall in 2010 (a) and 2011 (b)

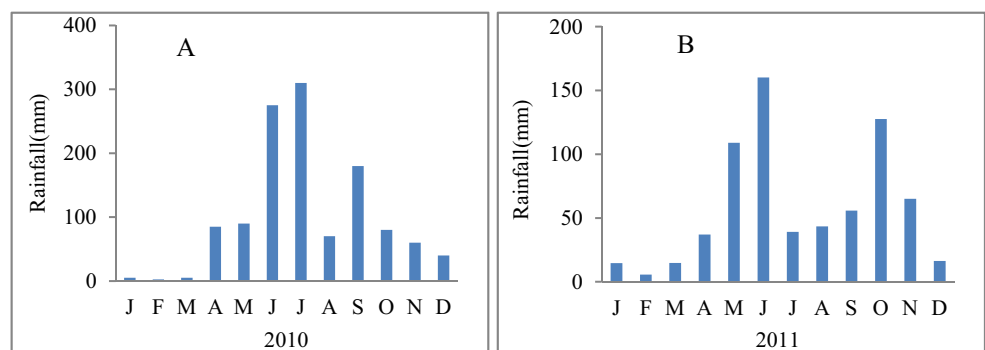
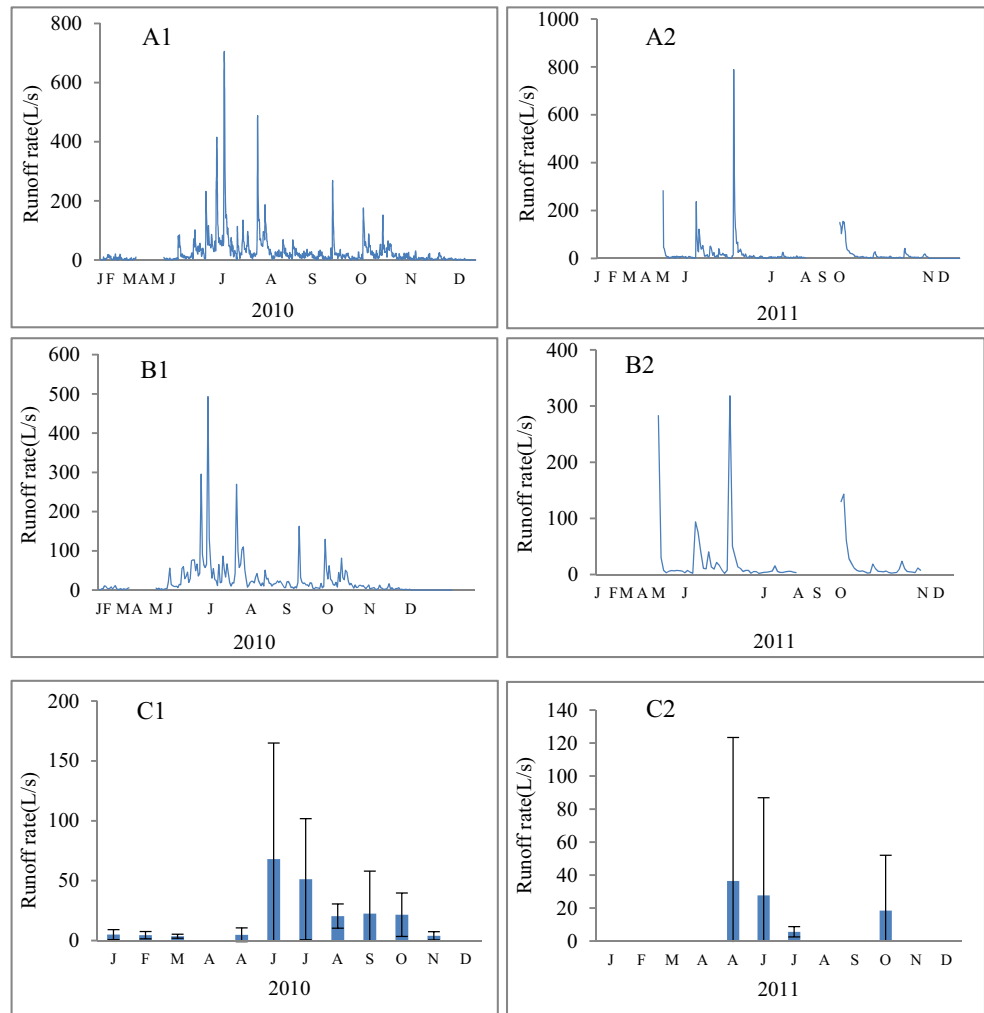


Fig. 5 Runoff processes shown by all the observed runoff rates (a(1) $N=44,436$ in 2010; a(2) $N=26,685$ in 2011), the daily average runoff rates (b(1) $N=211$ in 2010; b(2) $N=84$ in 2011), and the monthly average runoff rates (c(1) $N=10$ in 2010; c(2) $N=4$ in 2011)



monthly average runoff rates also showed high degrees of variation among different months in 2010 and 2011. Standard deviations of runoff rates for each month were greater than the averages, indicating a non-normal distribution in runoff rates (Fig. 5c(1), c(2)).

TN

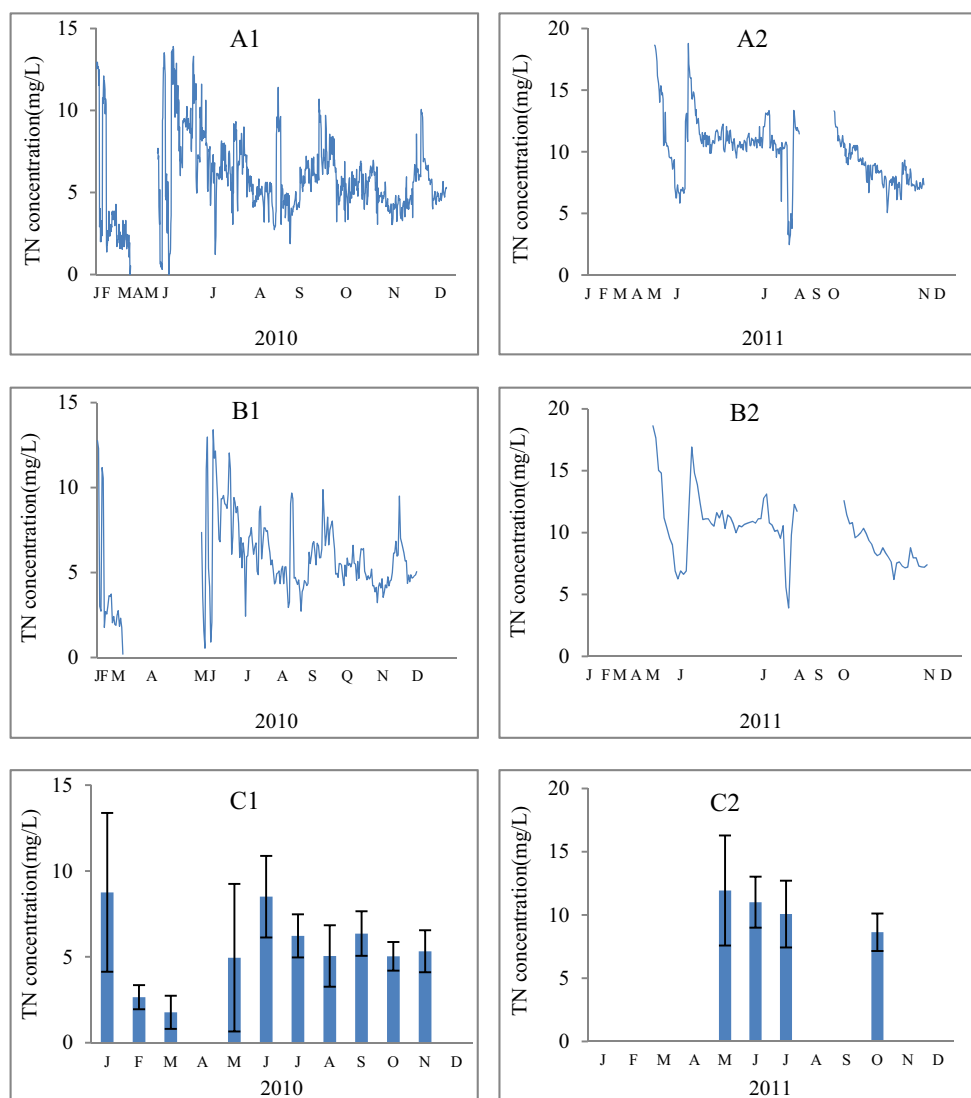
We collected 1085 runoff samples in 2010. The TN concentrations in these runoff samples changed from 0.06 to 14.5 mg/L and averaged 6.17 mg/L (Fig. 6a(1)). The TN concentrations in 98.71% and 95.85% of the runoff samples exceeded the upper limits of the TN concentrations in the third and fifth types of surface water, respectively, according to the Environmental Quality Standards for Surface Water (EQSSW) in China (Supplemental material 2). The number of runoff samples was 438 in 2011. The TN concentrations in these runoff samples changed from 2.47 to 18.79 mg/L and averaged 12.12 mg/L (Fig. 6a(2)). The TN concentrations in all the runoff samples exceeded the upper limit of TN

concentration in the fifth type of surface water. The runoff flowing into Lake Hongfeng from the small Dachong watershed was seriously polluted by nitrogen. The TN concentration in the surface runoff during the 2011 drought year was significantly higher than that in the normal rainfall year of 2010 (Fig. 6a(1)–c(1), a(2)–c(2)). The variation in TN concentrations within each month was relatively small based on the standard deviation (Fig. 6c(1), c(2)).

TP

The TP concentrations in the surface runoff in 2010 ranged from 0.01 to 1.57 mg/L and averaged 0.2 mg/L (Fig. 7a(1)). The TP concentrations in 95.76% of the runoff samples were lower than the upper limit of TP concentration in the third type of surface water according to the EQSSW in China (Supplemental material 2). Only 4.24% of the runoff samples were unqualified. The TP concentrations in the runoff samples in 2011 ranged from 0.01 to 0.59 mg/L and averaged 0.22 mg/L (Fig. 7a(2)). The TP concentrations in 91.19% of the runoff

Fig. 6 TN concentrations in each of the runoff samples monitored through the years 2010 (**a(1)** $N=1085$) and 2011 (**a(2)** $N=438$), the daily average TN concentrations (**b(1)** $N=211$ in 2010; **b(2)** $N=84$ in 2011), and the monthly average TN concentrations (**c(1)** $N=10$ in 2010; **c(2)** $N=4$ in 2011)



samples were lower than the upper limit of the TP concentration in the third type of surface water according to the EQSSW in China. The percentage of the unqualified runoff samples was 8.81%. The runoff flowing into Lake Hongfeng was not obviously polluted by phosphorus. The TP concentration in the runoff in 2010 was generally higher than that in 2011 (Fig. 7a(1)–c(1), a(2)–c(2)). The TP concentrations showed a relatively greater variation based on their standard deviation than the TN concentrations (Figs. 6a, c(1), c(2) and 7c(1), c(2)).

COD

The COD concentrations in the runoff samples in 2010 ranged from 0.01 to 160.00 mg/L and averaged 26.29 mg/L (Fig. 8a(1)). COD concentrations in 41.43% and 19.53% of runoff samples were higher than the upper limits of the COD concentrations in the fifth and third types of surface water, respectively. The COD concentrations in the runoff samples

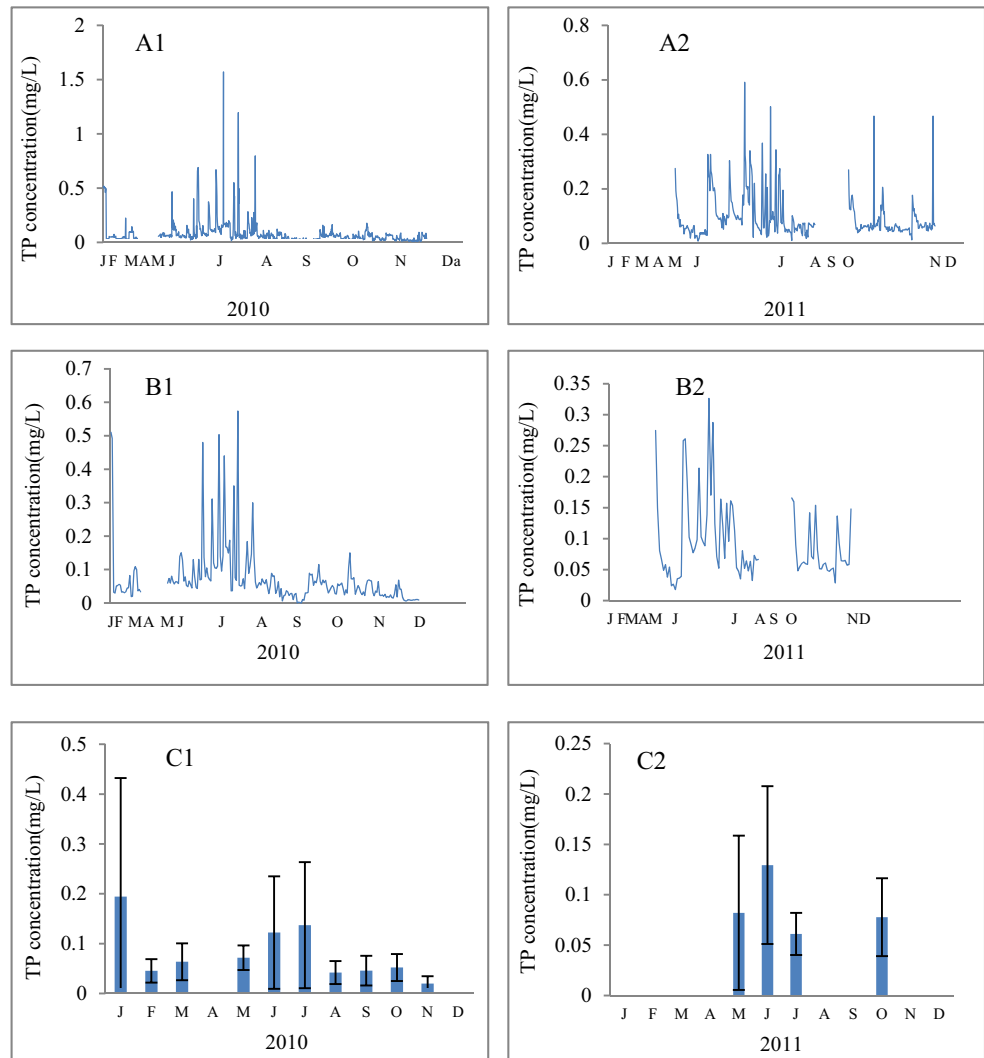
in 2011 ranged from 12.00 to 140.00 mg/L and averaged 44.55 mg/L (Fig. 8a(2)). The COD concentrations in 77.38% and 22.62% of the runoff samples were higher than the upper limits of COD concentration in the third and fifth types of surface water, respectively. The surface runoff from the small Dachong watershed was seriously polluted by organic pollutants. The variation in the COD concentrations during February 2010 and May 2011 was relatively greater than that during the other months (Fig. 8b(1), b(2)).

Occurrence frequencies of different runoff rates and TN or TP concentrations

Occurrence frequencies based on the values of all observations

The small runoff rates occurred at a high frequency both in 2010 and 2011, but the large runoff rates occurred at a very

Fig. 7 TP concentrations in each of the runoff samples monitored through the years 2010 (a(1) $N=1085$) and 2011 (a(2) $N=438$), the daily average TP concentrations (b(1) $N=211$ in 2010; b(2) $N=84$ in 2011), and the monthly averages (c(1) $N=10$ in 2010; c(2) $N=4$ in 2011)



low frequency (Fig. 9a(1), a(2)). The occurrence of runoff events did not belong to normal, uniform, exponential, or Poisson distributions ($N=44,436$, $p < 0.000$; $N=26,685$, $p < 0.000$; Supplemental material 3), although the histograms were similar to Poisson distribution forms. However, a moderate concentration of TN in the surface runoff occurred at a high frequency both in 2010 and 2011, and high and low concentrations of TN occurred at a low frequency (Fig. 9b(1), b(2)). The Kolmogorov-Smirnov test indicated a normal distribution form ($N=1085$, $p=0.198$; $N=438$, $p=0.105$). The occurrence frequency of different TP concentrations in the surface runoff was similar to that of different runoff rates per the histograms (Fig. 9c(1), c(2)), but their distribution did not belong to any one of the four forms ($N=1085$, $p=0.000$; $N=438$, $p=0.017$).

Changes of the frequencies based on the daily averages

The occurrence frequency of the daily average runoff rates and TN and TP concentrations in 2010 and 2011 showed

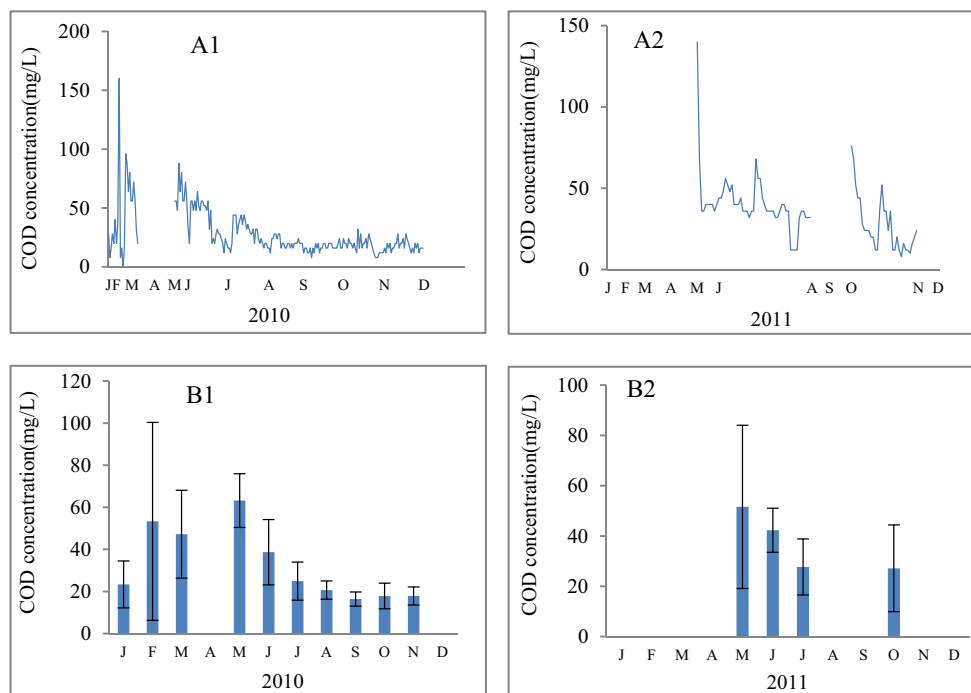
some changes when the data of all observations were averaged (Fig. 10). However, the distributions of the daily average runoff rates still did not belong to the four forms identified ($N=365$, $p=0.034$; $N=365$, $p=0.000$; Supplemental material 3). The daily average TN concentrations were obviously normal in their distribution forms ($N=1085$, $p=0.198$; $N=438$, $p=0.105$). The TP concentrations trended to a normal distribution from the previously described form ($N=211$, $p=0.214$; $N=211$, $p=0.004$). The occurrence of different COD concentrations in 2011 also presented a normal distribution based on the Kolmogorov-Smirnov test ($N=211$, $p=0.062$).

Runoff quantity and TN, TP, and COD loss

Monthly runoff quantity and TN, TP, and COD loss

Runoff quantity in the second half of year 2010 was far greater than that of the first half of year because it rained a large amount from June to October (Fig. 11a(1)). Fertilizers were often applied to croplands in these months. Consequently, the

Fig. 8 Daily averages and monthly averages of COD concentrations in the runoff samples. The daily averages in **a(1)** 2010 ($N=211$) and **a(2)** 2011 ($N=84$) and the monthly averages in **b(1)** 2010 ($N=10$) and **b(2)** 2011 ($N=4$). For the tests of COD concentrations, all runoff samples collected during the day were mixed into a water sample; then, the COD concentration in the water sample was measured. Thus, only the daily and monthly averages of the COD concentrations were obtained over the 2 years. COD_{cr} in the figures represents the test results with dichromate titration, and the same below abbreviation is below



loss of TN, TP, and COD with surface runoff was high, which accounted for 94.8%, 73.3%, and 94.3% of the total TN, TP, and COD losses, respectively (Fig. 11b(1), c(1), d(1)). However, rainfall was very minimal from February to May in 2010, and all paddy fields in the watershed were under dry farming conditions. Correspondingly, the runoff quantity was small (Fig. 11a(1)). The loss of TN, TP, and COD with surface runoff only accounted for a small percentage of the total loss. Although rainfall events in April and May were larger than those in August (Fig. 4a(1)), the cultivation and transplantation of rice seedlings consumed substantial surface runoff during these 2 months. Therefore, the runoff quantity and the loss of TN, TP, and COD with runoff were much less in April and May than in August. In September and October, there was substantial drainage from the rice field, which caused a large amount of runoff and loss of TN, TP, and COD (Fig. 11a(1)–d(1)).

Surface runoff primarily occurred in June and October of 2011 because severe droughts occurred in other months (Fig. 11a(2)). Correspondingly, the runoff quantity of the 2 months accounted for 86.42% of the total runoff. The losses of TN, TP, and COD with the runoff were also high during those 2 months, and the losses were as high as 83.78%, 80.56%, and 81.95%, respectively, during the whole year (Fig. 11b(2), c(2), d(2)).

Annual runoff quantity and TN, TP, and COD loss

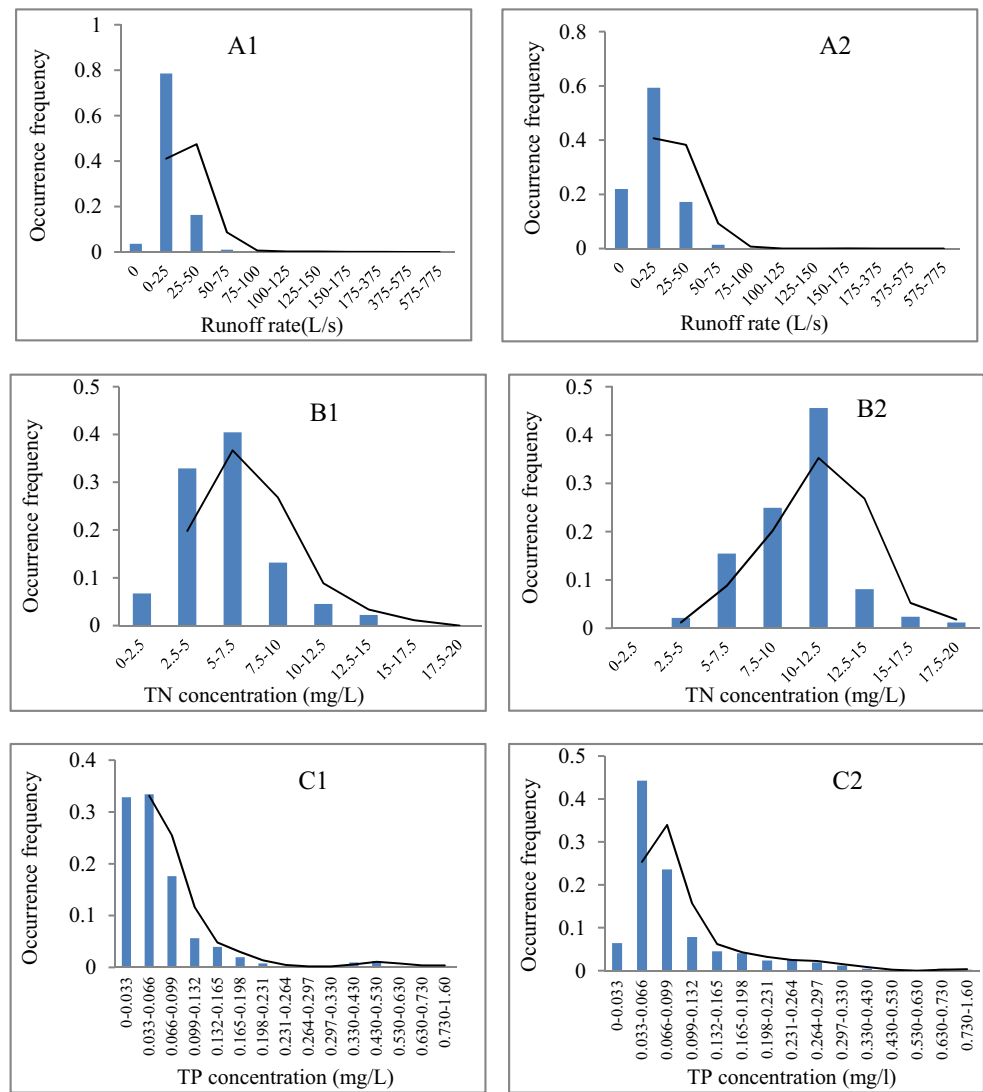
Runoff quantity and the loss of TN, TP, and COD with the surface runoff in 2010 were significantly higher than

those in the 2011 drought year (Table 3). Compared with 2010, in 2011, the runoff quantity and the loss of TN, TP, and COD with the surface runoff decreased by 64.73%, 30.06%, 61.57%, and 39.85%, respectively. Runoff and TP showed a relatively obvious decrease among these four indices.

Relationships between runoff rates and concentrations or loss of TN, TP, or COD

There were no significant relationships between runoff rates and TN concentration in the surface runoff in 2010 and 2011 (Fig. 12a(1), a(2)). The TN concentration showed a slight increase with increasing runoff rates at low runoff rates but decreased at high runoff rates. The 2010 normal rain year and the 2011 drought year were highly similar in their relationships. However, there were statistically significant relationships between runoff rates and TP concentration in surface runoff, although statistical significance was not high (Fig. 12b(1), b(2)). The relationships between the runoff rates and COD concentration in the surface runoff in 2010 and 2011 showed different statistically significant results (Fig. 12c(1), c(2)). The loss of TN, TP, or COD with the runoff in 2010 and 2011 showed identically significant relationships with increasing runoff rates (Fig. 12d(1)–f(1), d(2)–f(2)). The relationships between the loss of the three indices and runoff rates could be fitted by linear equations (Fig. 12d(1)–f(2)).

Fig. 9 Occurrence frequencies based on the values of all observations. **a(1), b(1), c(1)** The occurrence frequencies of different runoff rates (in 44,436 samples) and TN and TP concentrations (in 1085 samples), respectively, in 2010. **a(2), b(2), c(2)** The occurrence frequencies of different runoff rates (in 26,685 samples) and TN and TP concentrations (in 438 samples), respectively, in 2011

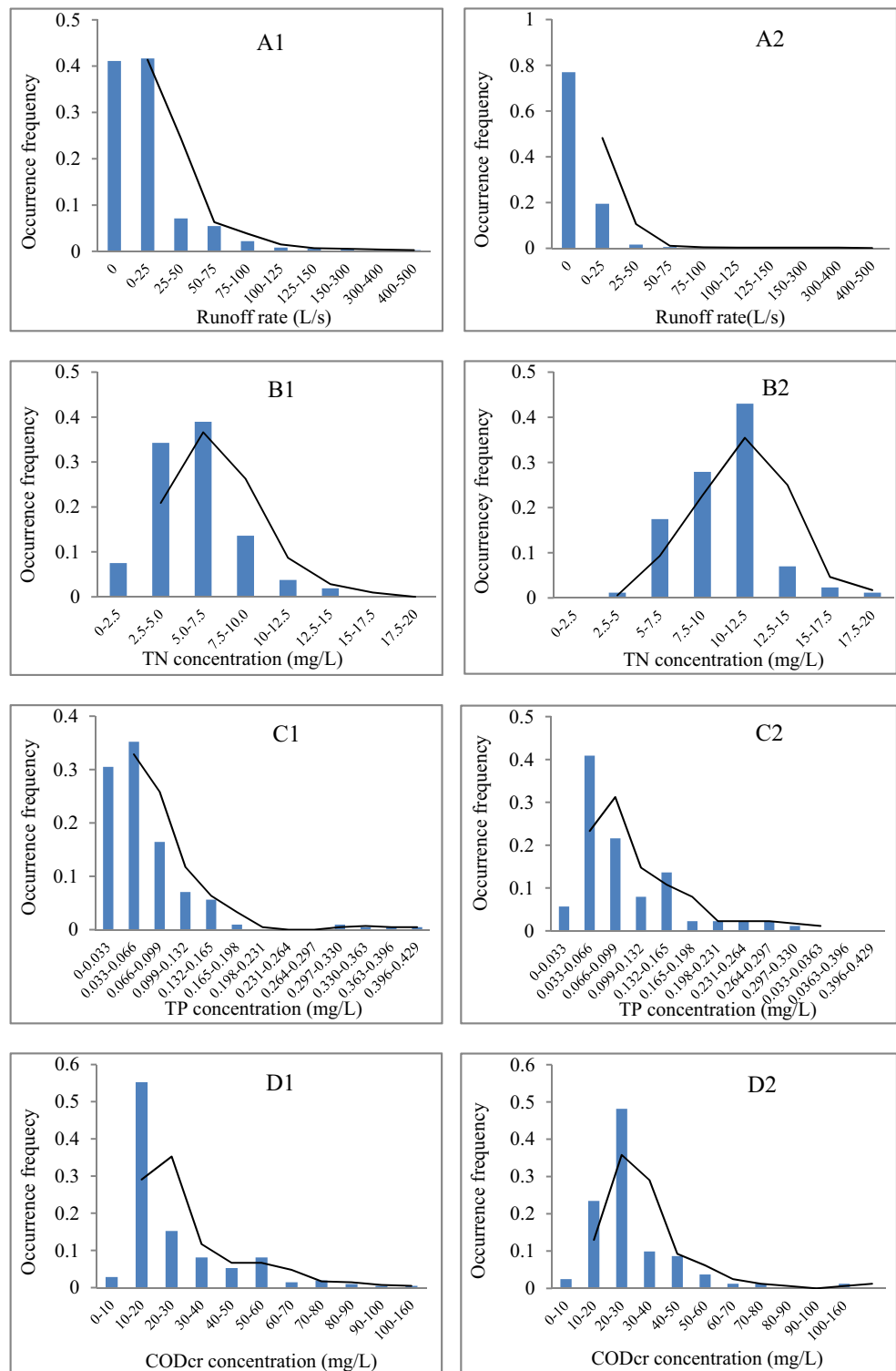


Discussion

The dynamics of runoff rates and N and P concentrations indicate the occurrence patterns and export behaviors of N and P in runoff from a watershed and allow a better understanding of how to control and treat pollutants in receiving waters (Surbeck 2005). Dynamic data of runoff rates and N and P concentrations may also be shared with local governments to determine if pollutants in runoff have exceeded established water quality standards for management of drinking-water source protection areas or to facilitate the establishment of new water quality standards. The results of the study indicated that runoff events occurred for 211 days and 84 days during the normal and drought rainfall years 2010 and 2011, respectively, in the small Dachong watershed. There was a great difference between the daily or monthly average minimum and maximum runoff rates. The TN concentrations in the runoff changed from 0.01 to 18.79 mg/L during the

2 years and averaged 7.06 mg/L. Almost all (i.e., 98.98%) of the runoff samples contained higher concentrations of TN than the upper limit (1 mg/L) of TN concentration in the third type of surface water. The TN concentration required an upper limit in surface water in the drinking-water source protection areas as per the EQSSW in China (Ministry of Environmental Protection of China 2002b). These test values of TN concentrations in the runoff over 2 years are obviously larger than the TN concentrations in the surface runoff from 58 ha of the Mkabela catchment covered by a variety of crops (Berry 2005) and from the plots in Mediterranean vineyards in Portugal (Ferreira et al. 2018). For the ranges of TP concentrations in the runoff samples collected during 2010 and 2011, only 5.71% of the runoff samples contained higher concentrations of TP than the upper limit (0.2 mg/L) of TP concentration in the third type of surface water, which met the requirement of the EQSSW in China for drinking-water source protection areas. Comparatively, TP concentrations (> 0.5 mg/L)

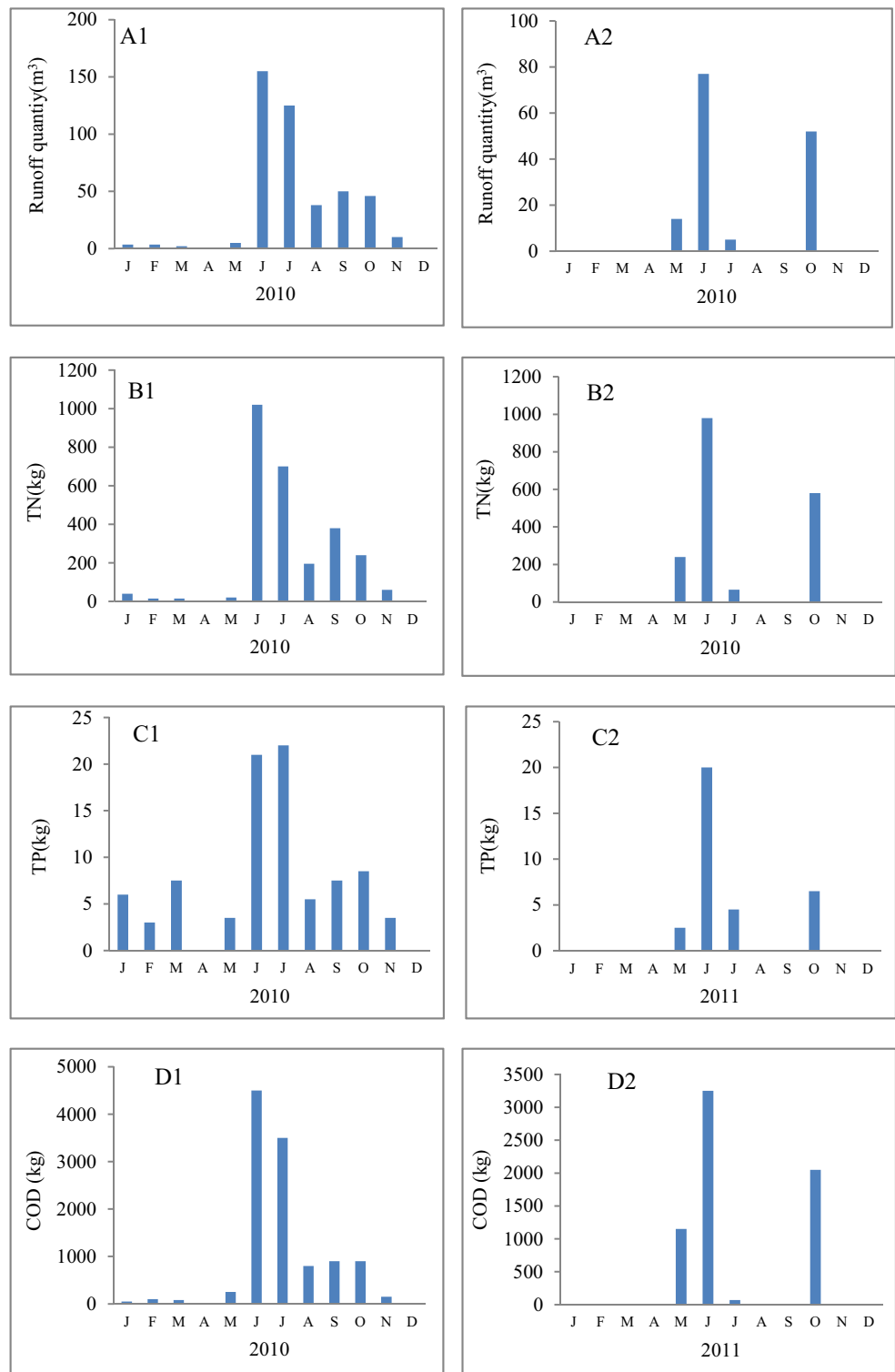
Fig. 10 Occurrence frequencies based on the daily averages of all observed values. **a(1), b(1), c(1), d(1)** The occurrence frequencies of different runoff rates (in 365 samples), TN and TP concentrations (in 211 samples), and COD concentrations (in 210 samples), respectively, in 2010. **a(2), b(2), c(2), d(2)** The occurrence frequencies of different runoff rates (in 365 samples), TN and TP concentrations (in 84 samples), and COD concentrations (in samples 84), respectively, in 2011



in most of the runoff samples collected from the plots in Mediterranean vineyards in Portugal during 2013 and 2014 were far higher than the test results in the study (Ferreira et al. 2018). The relatively lower concentrations of TP and available phosphorus in the soil of a karstic region than in a nonkarstic region may explain the low concentrations of TP in the runoff

(Zhang et al. 2014; Table 2). Thus, the runoff flowing into Lake Hongfeng was not obviously polluted by phosphorus. The COD concentrations in the runoff samples during 2010 and 2011 ranged from 0.01 to 160.00 mg/L and averaged 29.95 mg/L. These results are far higher than the observed values in the nonkarstic watershed of Lake Youngrang located

Fig. 11 Runoff quantity and TN, TP, and COD losses from the small Dachong watershed. **a(1)**, **b(1)**, **c(1)**, **d(1)** Runoff quantity and TN, TP, and COD losses, respectively, in 2010. **a(2)**, **b(2)**, **c(2)**, **d(2)** Runoff quantity and TN, TP, and COD losses, respectively, in 2011



on the eastern coast of South Korea (Jaeheon 2013). More than half (i.e., 52.04%) of the runoff samples during 2010 and 2011 were higher than the upper limit (20 mg/L) of COD concentration in the third type of surface water according to the EQSSW in China. Land use includes croplands, forests, gardens, and residential areas in the Mkabela

catchment and the watershed of Lake Youngrang (Berry 2005; Jaeheon 2013). The small Dachong watershed also includes these types of land use and orchards to achieve an economic goal, similar to Mediterranean vineyards in Portugal (Ferreira et al. 2018). Rainfall in the three sites is similar to that in the Dachong watershed. However, the TN

Table 3 Runoff quantity and loss of TN, TP, and COD in 2010 and 2011

Time	Runoff quantity		TN		TP		COD	
	Total (m ³)	Unit load (m ³ /ha/year)	Total (kg)	Unit load (kg/ha/year)	Total (kg)	Unit load (kg/ha/year)	Total (kg)	Unit load (kg/ha/year)
2010	4.14 × 10 ⁵	3341.39	2546.7	20.55	85.6	0.69	10,883.2	87.86
2011	1.46 × 10 ⁵	1178.37	1781.0	14.38	32.9	0.26	6546.7	52.85

or COD concentrations in the runoff from the small Dachong watershed far exceeded those from the three sites that do not belong to the drinking-water source protection area. The small Dachong watershed is a representative sample of the drinking-water source protection area of Lake Hongfeng. Lake Hongfeng provides 70% of the drinking water for citizens

living in Guiyang City. It is urgent to control the pollution of nitrogen and COD in surface water for these citizens.

With a further analysis of the runoff rates and N and P concentrations, we determined the probability distribution of the occurrences of different runoff rates and N and P concentrations in the small Dachong watershed. Rainfall at different

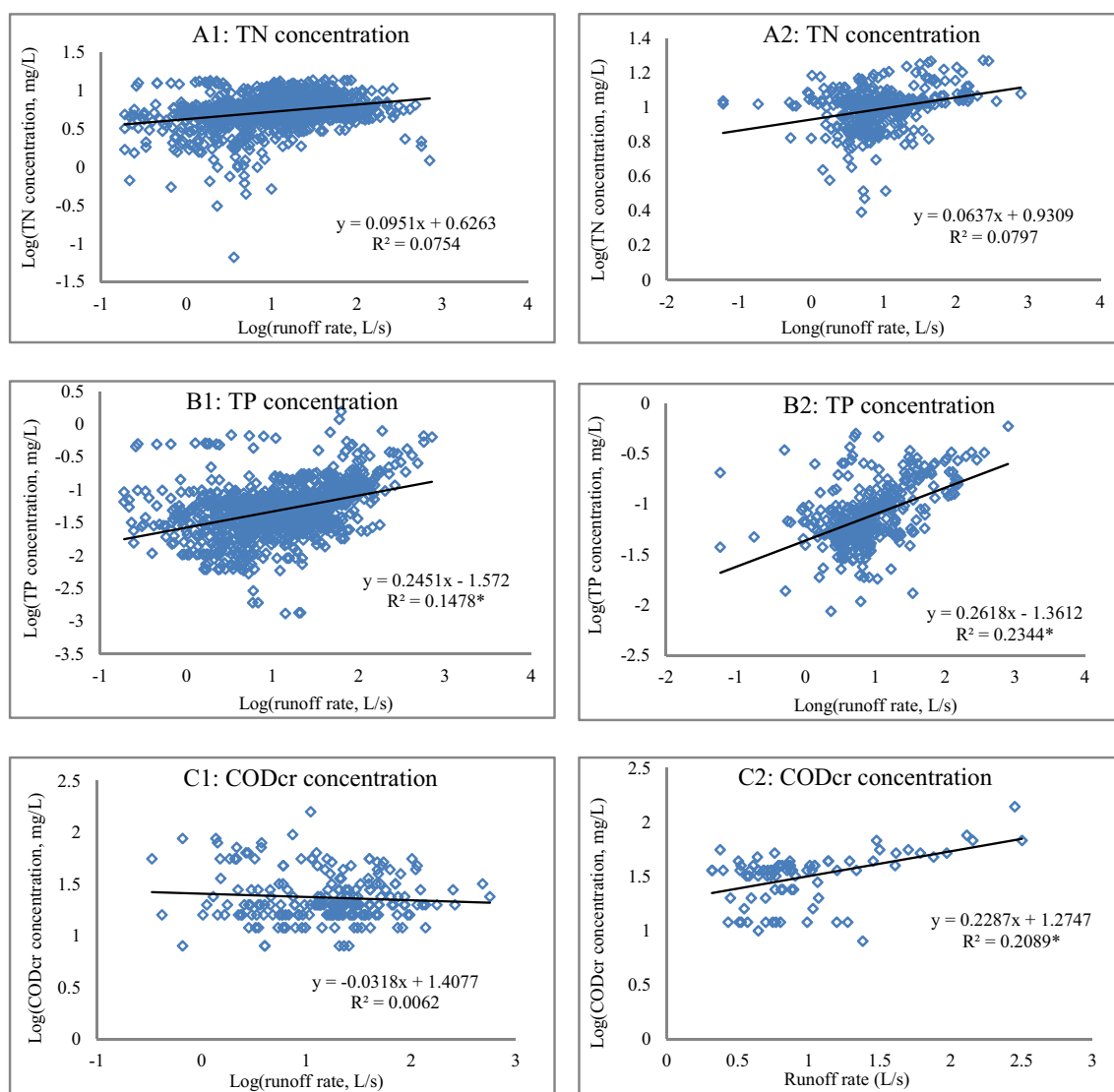


Fig. 12 Relationships between runoff rates and concentrations and losses of TN, TP, or COD in runoff by logarithmic transformation. Concentrations of TN, TN, or COD in runoff were the test values at the moment runoff rates were tested; corresponding loss was a total of TN,

TN, or COD in the runoff for 3 h. **a(1)–f(1)** The analysis results based on 1085 runoff samples in 2010. **a(2)–f(2)** The analysis results of 438 runoff samples in 2011. Single, double, and triple asterisks represent statistical significance at the 95%, 99%, and 99.9% confidence levels, respectively

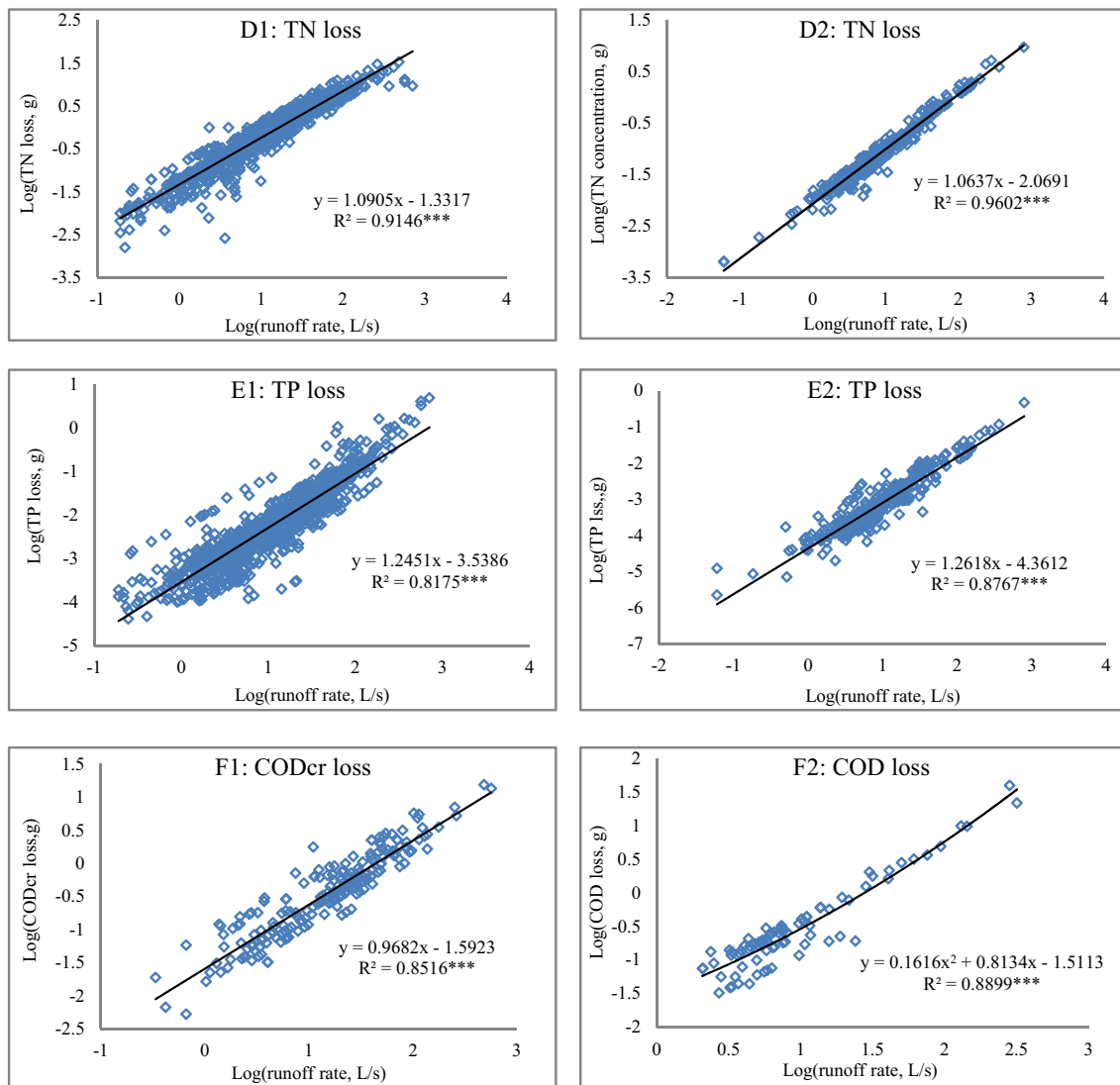


Fig. 12 continued.

times is a random event, and correspondingly, different runoff rates and N and P concentrations in the surface runoff caused by rainfall are random events. The occurrence frequency of a random event can show the occurrence law of the random event to effectively manage or avoid that random event (Núñez et al. 2011). We found that low surface runoff rates often occurred at a high frequency, and high surface runoff rates occurred at a very low frequency. The occurrence frequency of different runoff rates presented some characteristics of the Poisson distribution, while the Kolmogorov-Smirnov test was not statistically significant (Figs. 9a(1), a(2) and 10a(1), a(2)). The occurrence frequency of different concentrations of TP is similar to that of the runoff rates. Previous results have indicated that particle-bound phosphorus is a major fraction of TP in surface runoff (Wendt and Corey 1980; Uusitalo et al. 2001). Organic matter and fine soil particles in topsoil and applied fertilizer and manure can be rapidly dissolved into rainfall runoff and become the primary sources of

particle-bound phosphorus in surface runoff (Buda et al. 2009). Compared with low surface runoff rates, high runoff rates cause more organic matter and fine soil particles to be transported with surface runoff (Wendt and Corey 1980). Thus, higher TP concentrations in surface runoff in the small Dachong watershed occurred in connection with high-rate runoff events because of the similar occurrence frequency of TP concentrations to that of surface runoff rates (Figs. 9c(1), c(2) and 10c(1), c(2)). However, the occurrence frequency of different concentrations of TN showed a normal distribution (Figs. 9b(1), b(2) and 10b(1), b(2)). Nitrogen in surface runoff is primarily in the form of dissolved nitrogen (Udawatta and Motavalli 2006). A moderate rate of surface runoff resulted in more nitrogen being dissolved from the soil, but a high rate of surface runoff might have diluted the nitrogen in the surface runoff. Thus, the occurrence frequency of moderate concentrations of TN in the surface runoff was predominant. The occurrence frequency of COD concentrations also had a

normal or close to normal distribution (Fig. 10d(1), d(2)). There is a similar explanation for the distribution of the COD concentrations as that of the TN concentrations (Jaeheon 2013). It is crucial for controls of TP in the surface runoff in the drinking-water source protection areas of Lake Hongfeng to highlight a high rate of surface runoff but, for controls of TN and COD, to highlight a moderate rate of surface runoff based on these occurrence frequencies.

We calculated the load of TN, TP, and COD loss with surface runoff by using the runoff rate and pollutant concentration data. The loads of TN, TP, and COD loss from the Dachong watershed in the 2010 normal rainfall year were 20.55 kg/ha/year, 0.69 kg/ha/year, and 87.86 kg/ha/year, respectively, but in the 2011 drought year, these values were only 14.38 kg/ha/year, 0.26 kg/ha/year, and 52.85 kg/ha/year, respectively. The loads for 2010 for the three indices were 1.5–2.5 times higher than those for the drought year. The results in the normal rainfall year showed loads higher than 13.5 kg/ha/year and 0.40 kg/ha/year of TN and TP loss, respectively, from the Chengjiang watershed in the nonkarstic region of the Three Gorges Reservoir (Yuan and Mu 2010) and 7.6 kg/ha/year of COD loss from the Lake Baiyang watershed (Sun et al. 2010). The annual mean rainfall in the Dachong watershed is similar to that in the Chengjiang watershed but higher than the 552.7 mm in the Lake Baiyang watershed. The productivity level of agriculture in the Dachong watershed is also analogous to that in both the Chengjiang and Lake Baiyang watersheds. Additionally, there is generally a relatively large background value of TN in the surface water from karstic watersheds compared with nonkarstic watersheds. Correspondingly, the loads of TN loss with the surface runoff from two small watersheds were only 0.15 kg/ha/year and 1.06 kg/ha/year, which were also far lower than the Dachong watershed (Lu et al. 2008). Such a low load of TN loss in the two watersheds is primarily due to low background values or low rainfall and runoff rates. However, in the nonkarstic Taihu watershed and Xinjiang River watershed of Southern China, which has high rainfall, there is a relatively high level of agricultural production and fertile soil, and consequently, the loads of TN losses from the two watersheds were as high as 44.53 kg/ha/year and 180 kg/ha/year, respectively, which are two times higher than those from the Dachong watershed (Li et al. 2004; Liu et al. 2017).

The results of the loads of TN, TP, and COD loss also provided basic data for determining the roles of different types of land use in N and P exports from the Dachong watershed. Zhang (2012) conducted observations of TN and TP losses with surface runoff from representative sites of SC, SEBF, SYF, and CFP in the Dachong watershed. It was assumed that surface runoff from these sites would flow out the watershed. Thus, the total loss of TN and TP from these types of land use was divided by the total loss of TN and TP from the mouth of the Dachong watershed observed in the study

(Table 3). Then, the roles of the four types of land use in N and P exports from the Dachong watershed were quantified. Specifically, SC, SEBF, SYF, and CFP contributed 7.5%, 5.5%, 6%, and 0.77%, respectively, of TN loss and 9.2%, 24.04%, 11.4%, and 4.74%, respectively, of TP loss to the total loss at the mouth of the Dachong watershed. These values are higher than the real contributions to total loss because a respectable amount of nitrogen and phosphorus transported from the four types of land use remained in the watershed (Berry 2005). Thus, these results highlight that other types of land use, such as VLs, PFs, TFs, and residential sites, had greater contributions to the total losses of TN and TP from the Dachong watershed than those of SC, SEBF, SYF, and CFP. Notably, SC only accounted for 5.1% of the area of the watershed, but its role in the total loss of TN was 7.5%, which was more than the roles of the larger SEBF and SYF in the area (Table 1). TN loss from SC in the Dachong watershed was analogous to that of a karst watershed in the middle section of Yunnan Province in China (Wang et al. 2009). The larger roles of VLs, PFs, TFs, and residential sites than the other types of land use in the total loss of TN were also found in nonkarstic regions (Li et al. 1995; Yu et al. 2005; Zhu 2008). Therefore, effective controls of N and P losses from VLs, PFs, TFs, SCs, and residential sites are needed for a healthy Hongfeng Lake.

An analysis of the relationships between surface runoff rates and concentrations or losses of TN, TP, and COD in the surface runoff from a watershed can predict, to some degree, the variation in the concentrations or losses of TN, TP, and COD only by observation of the surface runoff at the mouth of the watershed (Chun et al. 2010). The differences in these relationships can also provide implications for management strategies for reducing the TN, TP, and COD in runoff (William and Michael 2011). Our observations indicated that there were no significant relationships between runoff rates and TN concentrations in surface runoff over the 2 years or COD concentration during 2010 (Fig. 12). TN and COD concentrations in surface runoff even decreased with increasing runoff rates, which might be due to TN and COD being diluted with increasing surface runoff rates. Correspondingly, a strategy for reducing the TN and COD concentration in surface runoff cannot be only dependent on the reduction in runoff rates. However, there were significant relationships between runoff rates and TP concentration and loss in surface runoff. The significant levels of the relationships between runoff rates and TN and TP losses in the Dachong watershed were obviously higher than those in the watershed in South Korea, where land use and rainfall were similar to those in the Dachong watershed (Chun et al. 2010). Therefore, a reduction in runoff rates is an effective method to control both TP concentration and total loss with surface runoff. Differences in the relationships of TN, TP, and COD concentrations with runoff rates have been found in many studies in nonkarstic regions

(Berry 2005; Chun et al. 2010; Ferreira et al. 2018). Thus, the results of this study are supported.

Conclusion

Surface runoff seasonally occurs in the small Dachong watershed. TN and COD concentrations in the surface runoff fluctuate far more than the upper limit of TN concentrations required by the EQSSW in China. Surface runoff in the drinking-water source protection area has been seriously polluted by nitrogen and organic pollutants. The TP concentration in surface runoff during the 2 years of the present study was mostly lower than the upper limit of the TP concentration required by the EQSSW in China. Surface runoff is not obviously polluted by phosphorus. The occurrence frequencies of different runoff rates and TP and COD concentrations show different distributions, but TN concentrations show a typical normal distribution. TP concentration and TN, TP, and COD losses are significantly related to runoff rates. TN, TP, and COD losses primarily occurred in vegetable lands, rice fields, and residential sites. It is urgent to implement control measures for nitrogen and COD pollution in drinking-water source protection areas for the health of the citizens in Guiyang City.

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